Thermal conductivity of $YBa_2(Cu_{1-x}Zn_x)_3O_{7-y}$ single crystals

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The in-plane thermal conductivity $K(T)$ of $YBa_2(Cu_{1-x}Zn_x)_3O_{7-y}$ single crystals (YBCO:Zn) has been investigated in the temperature range from 5 to 250 K and in magnetic fields up to 30 kOe. The thermal conductivity decreases substantially with increase of the Zn concentration providing reduction in the thermal conductivity peak and its disappearance at a high content of the dopant. At temperatures below 15 K the $K(T)$ dependencies are functionally similar for all the samples. The magnetic field effect on the thermal conductivity is strongly suppressed by doping even in the crystal with the lowest Zn content. For YBCO:Zn with $x = 0.003$ at temperatures below 25 K the magnitude of remnant magneto-thermal resistivity (after switching off the 30 kOe magnetic field) is about one tenth of that for pure YBCO. Analysing experimental data we use the theory of thermal conductivity in high-T_c superconductors and speculations about the modification of quasiparticle excitation spectrum due to the Cooper pair breaking.

74.25.Fy, 74.60.Ec, 74.62.Dh, 74.72.Bk

I. INTRODUCTION

The results of many experimental works, e.g., tunnel[1] and Raman[2–4] spectroscopy studies indicate that there is nonzero electron density of states inside the energy gap in YBCO in the superconducting state. The appearance of these states can be due to the effects of the Cooper pair breaking,[5] the breaking mechanism being either intrinsic or the result of some structural defects that exist even in the high quality single crystals of YBCO synthesized until now. For example, Lee and Readon[6] have shown that strong inelastic scattering of electrons results in the Cooper pair breaking. However, the origin of high scattering rate of electrons is not known in high temperature superconductors (HTSC).

Investigations of thermal conductivity of HTSC revealed that the heat conduction in these materials is mainly due to phonons: quite rough estimations of the phonon and electron contributions give 2/3 and 1/3 of the total thermal conductivity, respectively.[7] Such situation is fortunate to study the electron-phonon interaction in HTSC by the thermal conductivity method. It is important that thermal phonons interact with electrons even at temperatures below the critical temperature T_c. At temperatures not very lower than T_c, the phonon scattering rate is determined essentially by the electron-phonon interaction. In it's turn, the phonon relaxation is the basic factor that determines the lattice thermal conductivity. The phonon scattering rate in electron-phonon processes depends upon the amplitude of electron-ion interaction, the mass of charge carriers, and the energy dependence of the electron density of states $N(E)$ within a layer of the order of k_BT near the Fermi level. The later is particular important for superconductors at $T < T_c$.

In the framework of a model of heat conduction that we follow, the finite density of the in-gap states must results in reduction of thermal conductivity in the superconducting state. In this connection, the investigation of thermal conductivity of YBCO under the controlled variation of the Cooper pair breaking rate is important.

For our opinion, for such investigations, the YBCO with partial substitution of copper by zinc is convenient. It is well known that even a small concentration of zinc decreases T_c considerably and, in the same time, makes unessential changes in lattice parameters.[8] Neutron and calorimetric measurements revealed that the phonon density of states in the low energy domain ($< 15 - 20$ meV)[9] and the Debue temperature T_D[10] does not practically change under zinc doping. This agrees with that the zinc valency in YBCO, its ionic radius, and atomic mass almost coincide with corresponding parameters of Cu^{2+}. From this one can expect that zinc impurity in YBCO lattice is not a point defect for phonons. On the other hand, according to the data on electric conductivity and Hall effect,[11] zinc is an effective center for electron scattering; and the doping produces unessential changes in the carrier concentration. Experimental data of the tunneling spectroscopy,[12] nuclear magnetic resonance[13] suggest the existence of quasi-particle in-gap states in the YBCO:Zn at $T \ll T_c$. This result is naturally explained under the assumption that zinc addition gives rise to the pair-breaking in YBCO.

Thus, comparing data on the thermal conductivity for pure YBCO, in which the intrinsic mechanism of pair

breaking exists, with YBCO:Zn, in which copper ions are partially substituted by zinc, the valuable information on the electron density of states in the superconducting state can be obtained. In this work, we present experimental data on the temperature and magnetic field dependencies of thermal conductivity for YBCO single crystals with partial substitution of copper by zinc and discuse the obtained results.

II. SAMPLES AND EXPERIMENTAL

Single crystals of YBCO:Zn were grown by the CuO flux method in Pt crucible.[14] The crystals were grown under flux cooling from 1150°C to 860°C in air. After this the crucible was removed from the furnace, the flux pour out, and the crystals were quenched. Typical dimensions of crystals were 1 – 3 mm in the *ab*-plane and 10 – 40 μm in the *c*-axis direction. Measurements of a chemical composition of samples using an electron microscope with X-ray microanalizator have shown that Pt concentration in YBCO:Zn crystals was smaller than 0.1 at.%. The single crystals were annealed in flowing oxygen at 600°C during 1 h following by the cool down to 300°C with rate 10°C/h. According to the data of X-ray structural measurements,[15] such annealing regime provides the oxygen content of 6.93 – 6.97 in YBCO. We did not measure the oxygen content in our crystals of YBCO:Zn. However, according to results of many measurements performed on ceramic samples of YBCO:Zn, the oxygen stoichiometry is independent (within an experimental error) upon zinc concentration up to $x = 0.1$ and is 6.97 ± 0.01 for annealing procedure similar to that we used (see, e.g., Ref. 16). From this we suppose that the oxygen content in the annealed crystals of YBCO:Zn is practically the same and near 7.0.

Using an optical microscope, we observed a well developed twinning structure on the crystals of YBCO:Zn. For thermal conductivity measurements we selected crystals without traces of the flux on their surface and with narrow superconducting transition width.

The transition of samples into the superconducting state was detected using the measurement of ac magnetic susceptibility (in magnetic field of 0.1 Oe at the frequency of 667 Hz) during the cool down from room temperature. Fig. 1 shows temperature dependence of magnetic susceptibility (a real part) for YBCO:Zn crystals, which we used for thermal conductivity measurements. It is seen that the width of superconducting transition is narrow for YBCO samples with small zinc concentration (0.4 K for pure YBCO and smaller than 1.4 K for samples with $x < 0.014$) and much broad for samples with $x > 0.02$. In Table 1, the dimensions, critical temperature, transition width (determined at the levels 0.1 and 0.9 of the maximum value of the magnetic susceptibility), and zinc concentration are given for samples under study. Zinc concentration in the samples with measured transition temperatures was determined using a microprobe chemical analyzer. The obtained data are shown in Fig. 2. A linear approximation of the data gives −13.5 K/at.% Zn for the rate of T_c variation, which agrees well with the data of Ref. 11 for single crystals of YBCO:Zn.

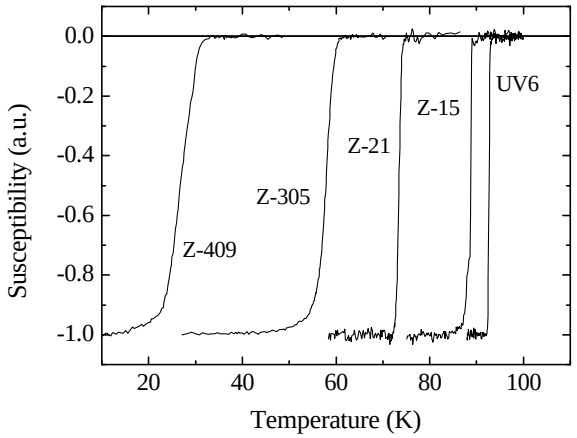

FIG. 1. Temperature dependence of the magnetic susceptibility for $YBa_2(Cu_{1-x}Zn_x)_3O_{7-y}$ single crystals in ac magnetic field $\mathbf{H} \parallel \mathbf{c}$, $H = 0.1$ Oe, $f = 667$ Hz.

TABLE I. Parameters of the $YBa_2(Cu_{1-x}Zn_x)_3O_{7-y}$ samples.

Sample	x	Dimensions, mm	T_c, K	ΔT_c, K
UV-6	0.000	$0.80 \times 0.30 \times 0.050$	92.8	0.4
Z-15	0.003	$0.90 \times 0.30 \times 0.040$	88.9	1.4
Z-21	0.014	$0.45 \times 0.30 \times 0.030$	74.0	1.2
Z-305	0.025	$0.45 \times 0.32 \times 0.014$	59.5	4.5
Z-409	0.047	$0.53 \times 0.38 \times 0.013$	30.0	7.0

The thermal conductivity was measured using a temperature-wave method at a frequency within the range from 0.2 to 2 Hz. The temperature wave in a sample was generated with an electrical heater glued at the end of the sample. The temperature difference along the sample (less than 2% of the mean temperature) was measured using a differential manganin-constantan thermocouple. In a separate experiment, we had determined a magnetic field dependence of the thermocouple sensitivity, and the values of thermal conductivity in magnetic fields were corrected. The statistical scatter of the data is about 0.1% and the systematic experimental error is 50%. The later is mainly due to the error in measured value of a distance between the thermocouple junctions, and in the heat current within the sample. The experimental technique will be published in more detail elsewhere.

III. RESULTS AND DISCUSSION

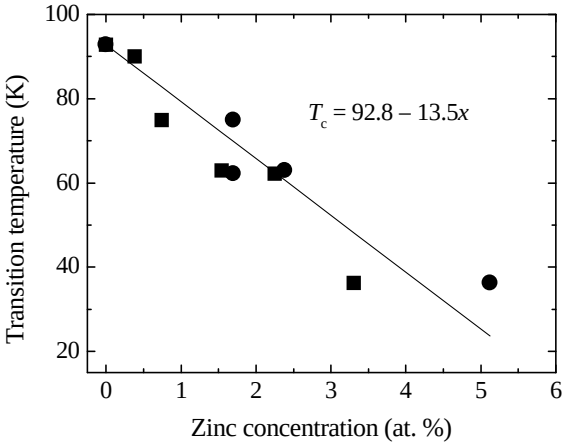

FIG. 2. Dependence of the superconducting transition temperature of $YBa_2(Cu_{1-x}Zn_x)_3O_{7-y}$ single crystals upon the atomic concentration of zinc. Circles and squares represent the data of two different measurements of zinc concentration, a solid line is a linear approximation of the data.

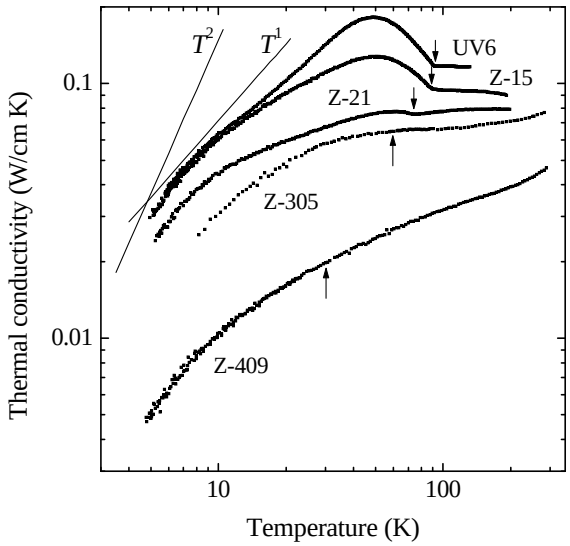

FIG. 3. Temperature dependence of thermal conductivity for $YBa_2(Cu_{1-x}Zn_x)_3O_{7-y}$ single crystals along the *ab*-plane. Arrows show the onset of superconducting transition.

A. Temperature dependencies of thermal conductivity

The temperature dependencies of the in-plane thermal conductivity for YBCO:Zn single crystals are shown in Fig. 3. With increase Zn concentration we observe: (1) a decrease of thermal conductivity in the temperature domain studied, this effect being stronger at low temperatures; (2) a decrease in relative magnitude of the maximum in thermal conductivity $K_{\max}/K(T_c)$ down to its disappearance for samples with $x > 0.025$.

In Ref. 17, we studied the temperature and magnetic field dependencies of thermal conductivity for YBCO single crystals. It was shown that experimental data can be quite well account for on the base of the theory,[18] which is the modification of the Bardeen, Rickaysen, and Tewordt (BRT) theory of thermal conductivity in superconductors[19] for the case of HTSC. We found that the phonon heat transport is a dominant mechanism of thermal conductivity. In YBCO, the electron-phonon interaction determines the behavior of the phonon thermal conductivity $K_{\rm ph}$ to a considerable extent. With temperature decrease below T_c, the electron thermal conductivity K_e decreases rapidly, because electrons condensed into the Cooper pairs and having a zero entropy do not participate in the heat transport, and the concentration of normal electrons decreases with a temperature. At the same time, the phonon relaxation due to the electron-phonon interaction decreases dramatically, resulting in the increase of $K_{\rm ph}$. The rise of the total thermal conductivity with temperature decrease continues until another mechanisms of the phonon relaxation (e.g., phonon scattering by crystal lattice defects, by sample boundaries, etc.) become dominant. At temperatures below the maximum, the thermal conductivity decreases mainly due to the decrease of population of thermal phonons; and $K(T)$ is determined mainly by the particular frequency and temperature dependence of the phonon relaxation rate.

Recently, Yu *et el.*[20] proposed a quite different scenario of the increase of thermal conductivity in YBCO at $T < T_c$. They attributed this feature to the decrease of quasiparticle relaxation rate in the superconducting state that gives rise to the dramatic increase in the electron thermal conductivity. Among basic experimental facts, which form the base of this model,[20] there is an absent of peculiarities in thermal conductivity of HTSC single crystals when the heat current is directed along the *c*-axis. This result was obtained by several groups. However, in the Ref. 21, authors observed the increase of the out-of-plane thermal conductivity in quite thin single crystals of yttrium and thallium cuprates. Therefore, we consider that the model of Yu *et el.*[20] has rather unsteady experimental foundations, and we will hold the phononic scenario. Until now, this (phonon) model explains all experimental data on the thermal conductivity of HTSC, at list qualitatively.

Proceed from such ideas on the heat transport, we qualitatively analyze the temperature dependence $K(T)$ of YBCO and its evolution upon copper substitution by zinc.

As noted above, in pure YBCO, the electron thermal conductivity is about 30% of the total at $T > T_c$. The observed considerable decrease of thermal conductivity of Zn-doped YBCO by factor of 2 – 4 for $x > 0.02$ cannot be a result of decrease in K_e only. Thus these experimental data indicate that $K_{\rm ph}$ in YBCO decreases considerably

with Zn doping. As the phonon scattering by lattice defects is not the dominant mechanism of the thermal resistivity in the temperature range considered, and the phonon-phonon scattering in YBCO:Zn does not change essentially due to weak sensitivity of lattice parameters and lattice dynamics to Zn impurities, then one can naturally assume that the observed effects to be a result of the increase in the rate of phonon-electron relaxation.

Previously, we have found[17] that, for the pure YBCO, the using of the following expression for the phonon-electron relaxation rate

$$\tau_{\text{ph-e}}^{-1} = A_{\text{ph-e}} x T, \tag{1}$$

where the relaxation amplitude has the form[22]

$$A_{\text{ph-e}} \approx (k_B/\hbar)(m^*)^2 E_{\text{def}}^2/(2\pi D\hbar^3 v_s), \tag{2}$$

gives quite reasonable values for parameters involved: $m^* \approx 16m_e$ is an effective electron mass (m_e is a free electron mass); $E_{\text{def}} \approx E_F \approx 0.3$ eV is a deformation potential, D is a specific density, and v_s is a sound speed. From the expression for $A_{\text{ph-e}}$ under the assumption that the deformation potential changes slightly with Zn doping, it follows that the effective electron mass increases, the increase being by factor of approximately two for $x = 0.047$.

In the normal state, the electron thermal conductivity K_{en} can be estimated by using the formula:[22]

$$K_{\text{en}} = \sigma(L_0 T), \tag{3}$$

$$\sigma = \frac{ne^2\tau}{m_{\text{ab}}}, \tag{4}$$

where σ is an electrical conductivity, n is a carrier concentration, e is an electron charge, m_{ab} is an effective electron mass in the ab-plane, τ is an electron relaxation time, and $L_0 = (k_B/e)^2\pi^2/3$ is the Lorenz number. On the base of these formulae one can expect the twofold reduction of the electrical and thermal conductivities of YBCO:Zn with $x = 0.047$ (in comparison with pure sample) as a result of increase of the effective electron mass, provided that this increase is totally due to m_{ab}. At $T > T_c$ the value of electron thermal conductivity is about 0.04 W/(cm K). According to the above presented analysis, the value of K_e must be smaller than 0.02 W/(cm K) and thus be less than 60% of measured thermal conductivity at 100 K. This magnitude is overestimated because zinc addition causes the decrease of the electron relaxation time.

The increase of effective electron mass should results in corresponding increase of a slope in the temperature dependence of electric conductivity (see Eq. 4). The authors of Ref. 11, which deals with measurements of in-plane $\rho(T)$ for single crystals of YBCO:Zn, have found that the slope $\mathrm{d}\rho/\mathrm{d}T$ increases by the factor of 1.7 as x increases from 0 to 0.035. This agrees with the increase of the mass m^* inferred from the value of the lattice thermal conductivity, which is estimated by using Eq. 2.

Let consider experimental data for low-temperature domain. In all samples of YBCO:Zn at temperatures below about 15 K, the dependencies $K(T)$ are functionally the same: they follow the T^1 dependence and change it to T^2 law at lowest temperatures.

Usually, in dielectric crystals at $T \ll T_D$, the phonon thermal conductivity is restricted by the sample dimensions due to phonon scattering at sample boundaries; and many other mechanisms of phonon relaxation, such as, for example, point defects, three phonon interactions, are ineffective.[23] In the boundary scattering regime, $K(T)$ should vary as T^3. But this does not agree with experimental data for single crystals of $YBa_2Cu_3O_{7-y}$[24] and $Bi_2Sr_2CaCu_2O_8$,[25] in which the quadratic dependence of $K(T)$ is observed at low and ultralow temperatures. In addition, the magnitude of thermal conductivity is much less than theoretical one. As suggested in Refs. 26,27, the mechanism, which gives rise to the T^2 dependence, is the phonon scattering by normal electrons. The existence of normal electrons at $T \ll T_c$ has evidently no connection with thermally excited quasiparticles above the gap, and is likely a consequence of finite density of states within the gap.

Let suppose that it is the phonon-electron interaction determines the low-temperature thermal conductivity of YBCO:Zn. As zinc concentration increases, the thermal conductivity of YBCO:Zn decreases slightly at $T < 15$ K (up to $x = 0.003$, $T_c = 88.9$ K), and then decreases much rapidly in comparison with the behavior at high temperatures. For example, at 6 K the thermal conductivity of the sample with $x = 0.047$ ($T_c = 30.0$ K) is 6.7 times smaller than the K(6 K) for pure YBCO, whereas at 100 K this ratio is about 3.7. This behavior of the low-temperature thermal conductivity indicates that, in the YBCO:Zn at $T \ll T_c$, the density of states within the layer of k_BT thickness at the Fermi level increases with zinc concentration.

We clarify how the change in the quasiparticle density of states $N(E)$ influences the low-temperature thermal conductivity. According to the theory of superconductors with paramagnetic impurities,[28,29] the pick in the $N(E)$ becomes broader with increase of dopant concentration and there is a finite density of states near the Fermi level at some concentration. For low impurity concentration and at low temperatures, the energy of thermal phonons is not enough high to excite quasiparticles. In this situation the phonon-electron scattering rate does not increase and the phonon thermal conductivity is practically the same as for pure superconductor. It is this we observe for the sample with $x = 0.003$. For higher impurity concentration, when $N(E)$ is finite at any energy, the phonon-electron relaxation channel works and this results in the decrease of phonon thermal conductivity even at very low temperatures. The essential decrease of the thermal conductivity of YBCO:Zn samples with relatively high zinc content agrees with this picture. In this

case, the allowable increase of the electron thermal conductivity does not realized because defects even of atomic scale strongly reduce K_e in metals, but the phonon thermal conductivity is weakly sensitive to points defects at $T \ll T_D$.[23]

In a number of works dealt with experimental investigation of the point contact spectra,[12] specific heat,[30] NMR and NQR on copper and yttrium nuclei,[13,31] there are conclusions about the existence of the gapless superconductivity in YBCO:Zn at $x > 0.05$ and about that the zinc impurity is an effective center for Cooper pairs breaking. The authors of Refs. 30,31 supposed that the pair breaking in YBCO:Zn has magnetic nature of the Abrikosov-Gor'kov type.[28] Note, the data of specific heat measurements in YBCO:Zn[30] indicate that there are in-gap excitations with practically zero energy even in samples with low zinc concentration ($x \approx 0.01$), for which the T_c decreases slightly. This result disagrees with the theory,[28] because it (theory) predicts the gapless superconductivity at relatively high pair-breaking rate when the critical temperature is much smaller than the initial "pure" value. On the other hand, in recently published theoretical work[5] dealt with pair-breaking effects in HTSC, the authors found such unusual behavior of the $N(E)$ in the superconducting state under strong inelastic pair-breaking scattering with even time-reversal symmetry.

Let now consider the experimental thermal conductivity data for intermediate temperatures but below T_c. With increase of zinc concentration the maximum in the thermal conductivity decreases and disappears for samples with $x > 0.02$. According to the theoretical models,[18,32] which agree well with experimental thermal conductivity data for YBCO,[17,33,34] the magnitude of thermal conductivity $K_{\max}$ in the maximum is determined mainly by the rate of phonon-electron relaxation and phonon scattering from lattice defects. The higher the rate of the normal electrons freeze out with temperature decrease, the higher $K_{\max}$. Evidently, if the $N(E)$ is nonzero within the gap then this results in the decrease of the $K_{\rm ph}$ and in some increase of the K_e. By the reasons mentioned above the gain in the total thermal conductivity from the increase of electronic component is apparently much smaller than the decrease of phonon component, and, as a result, the decrease of $K_{\max}$ is observed with zinc content rise. Thus, for intermediate temperatures, experimental data agree with the ideas about thermal conductivity behavior in superconductors with the Cooper pairs breaking.

In the literature, as we know, there are no theoretical works on the investigation of pair-breaking effects, e.g., caused by paramagnetic impurities, on the *phonon* thermal conductivity of superconductors. In Ref. 35, the theory of electron thermal conductivity of superconductors with paramagnetic impurities was developed. Our estimations of the phonon thermal conductivity for the case of finite density of states at the Fermi level show that the dependence of $K_{\rm ph}(T)$ is quite sensitive to the form of the function $N(E)$, which strongly varies over the energy range of the order of k_BT. This means that the calculations of the $K_{\rm ph}(T)$ requires the detail knowledge of the electronic spectrum of the HTSC.

B. Thermal conductivity in magnetic field

At $T < T_c$, the thermal conductivity of pure YBCO decreases in the magnetic field.[17,36] We interpret this as a result of the additional phonon scattering by normal electrons within the vortex cores.[17]

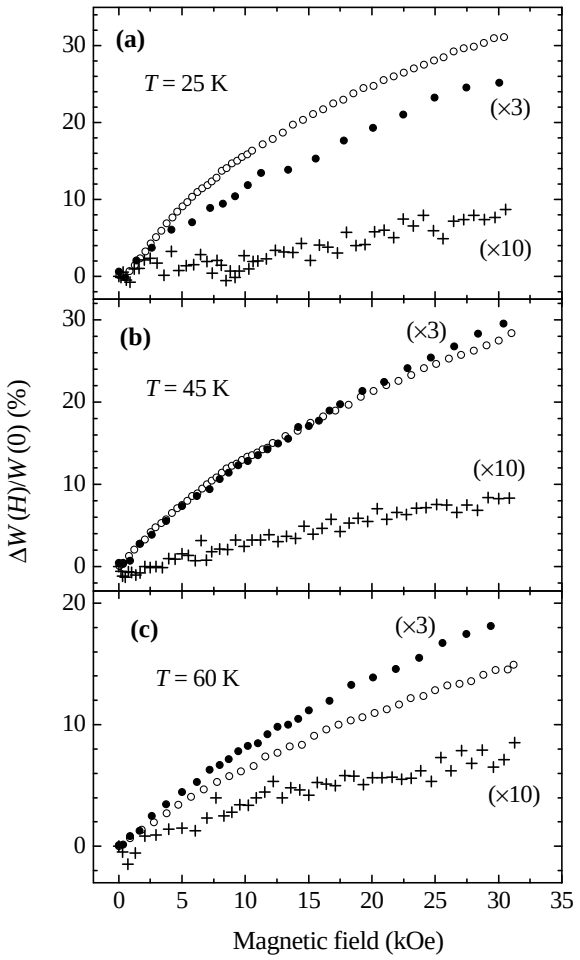

FIG. 4. Excess in-plane thermal resistivity vs magnetic field $\mathbf{H} \parallel \mathbf{c}$ for three $YBa_2(Cu_{1-x}Zn_x)_3O_{7-y}$ crystals at $T = 25$ K (a), 45 K (b), and 60 K (c). ∘ – sample UV-6, $x = 0.0$; • – sample Z-15, $x = 0.003$; + – sample Z-21, $x = 0.014$. The $\Delta W(H)/W(0)$ is multiply by factor of 3 and 10 for samples Z-15 and Z-21, respectively.

In accord with the supposition about the increase of $N(E)$ at the Fermi level in YBCO:Zn in the superconducting state, one can expect that the increase of the concentration of normal component in the magnetic field will

take place at the background of already existing density of states, and, thus, the higher doping level the weaker magnetic field effect. In Fig. 4, the field dependencies of the excess thermal resistivity $\Delta W(H)/W(0) = (K(H)^{-1} - K(0)^{-1})K(0)$ are shown for three YBCO:Zn samples with zinc concentration 0.0, 0.003, and 0.014 at temperatures 25 K, 45 K, and 60 K for magnetic field orientation parallel to the c-axis of a crystal. The magneto-thermal resistivity deceases with increase of zinc concentration: the effect smaller by factor of 2 – 4 and of 20 – 40 for $x = 0.003$ and 0.014, respectively. The suppression of magnetic field effect is larger at low temperatures. The obtained data generally agree with our expectations. Note, that the reduction of the magnetic field effect with zinc concentration correlates with the disappearance of the maximum in $K(T)$, because the phonon-electron interaction determines both of them.

At the same time, we note additional important (for our opinion) detail: in the sample of YBCO:Zn with $x = 0.003$, the lattice thermal conductivity is suppressed only slightly at $T < 25$ K as seen from temperature dependence of $K(T)$. Therefore, the essential suppression of the magnetic field effect on the thermal conductivity of YBCO:Zn must be determined by other factors. The origins of such behavior are not clear now. It is possible that the rate of phonon scattering by vortices depends upon the vortex pinning force. Possibly, in pure YBCO, the substantial increase of the thermal resistivity in magnetic field is connected with more strong pinning than in samples doped with zinc. The later conclusion can be inferred from the absence of the hysteresis phenomena in the YBCO:Zn samples. Moreover, our measurements of the magnetization showed that at $T = 25$ K the remnant magnetic moment $M_{\rm rem}$ for the sample YBCO:Zn with $x = 0.003$ is about two times smaller than for pure YBCO (see Fig. 5) and, therefore, the pinning force is smaller as well.

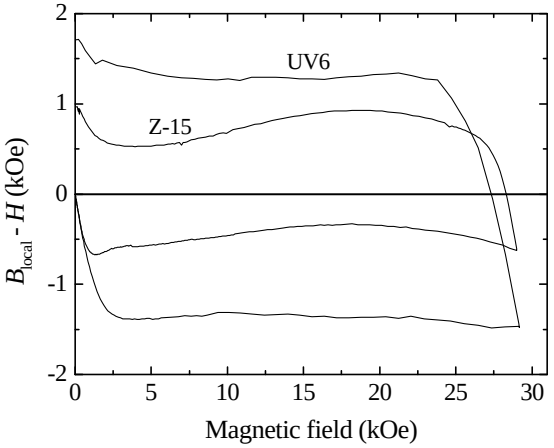

FIG. 5. Magnetization curves for pure YBCO (UV6) and $YBa_2(Cu_{0.997}Zn_{0.003})_3O_{7-y}$ (Z-15) in the magnetic field oriented parallel to the c-axis at $T = 25$ K.

IV. SUMMARY

For the first time we have measured the temperature dependencies of thermal conductivity for $YBa_2(Cu_{1-x}Zn_x)_3O_{7-y}$ single crystals in the temperature range from 5 to 250 K. The copper substitution by zinc results in essential reduction of thermal conductivity and in the decrease of thermal conductivity pick down to its complete disappearance for samples with zinc concentration above 0.025. At temperatures below 15 K, dependencies of $K(T)$ for different samples are functionally the same.

Analysis of experimental data shows that in YBCO the effective electron mass increases with Zn concentration rise, and the density of states within the gap (in the superconducting state) is nonzero even for smallest zinc concentrations. The later can not be account for in the frame of simple theory of superconductors with paramagnetic impurities.[28,29] This suggests that some unusual mechanism of Cooper pair breaking may exist in YBCO:Zn crystals, the similar mechanism being in pure YBCO and resulting in the quadratic temperature dependence of thermal conductivity at low temperatures.

Introduction of zinc into YBCO causes the essential suppression of the magnetic field effect on the thermal conductivity in the ab-plane in the case when the field is directed perpendicular to the heat current. In contrast to the case of pure YBCO, no hysteresis phenomena are observed in the thermal resistivity. The comparative analysis of experimental data for different YBCO:Zn samples suggests that the pinning of the vortex line may results in the increase of the thermal resistivity in magnetic field.

ACKNOWLEDGMENTS

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* This paper was published in Russian journal which is not easily accessible. The authors believe that the subject matter of the paper is steel interesting. Because of these, the authors have prepared the english version of the paper and placed it into the archive. Interpretation and discussion of

data may be now out of date in some aspects, but the experimental data seem to be the issue of the day.

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